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RADIATION LIFETIMES AND FAILURE MECHANISMS OF CARBON STRIPPER FOILS

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Abstract

Measurements of "lifetimes" of thin carbon foils under heavy-ion irradiation are compiled and recent advances in stripper foil technology are reviewed. The impact of recent foil lifetime improvements, many by more than an order of magnitude, on heavy-ion electrostatic accelerators is discussed. Foil inhomogeneities, particularly those caused by sputtering are suggested to be a prime factor in usable foil lifetimes.

Historical

The ever-increasing use of tandem electrostatic accelerators for producing intense heavy-ion beams has generated a great deal of concern regarding the usable lifetimes of stripper foils. The advantage of foil-stripping over gas-stripping is graphically illustrated[1] in fig. 1, which shows the fractional transmission of ^{32}S and ^{208}Pb beams for different stripper combinations. It is clear that the advantage of foil stripping over gas stripping increases rapidly as one goes to higher masses and projectile energies.

The drawback to using foil strippers became painfully apparent when it was found that the foils deteriorated rapidly under heavy-ion bombardment. Typical foil lifetimes of only a few minutes were indicated[2] which would have required frequent reloading of the foil holder. This is a costly and time consuming process and would have placed severe restrictions on accelerator operations. This underlines the importance of recent efforts toward the study and improvement of carbon foil lifetimes.

Until recently, carbon stripper foils had been manufactured almost exclusively by conventional vapor deposition techniques. Although attempts to improve the lifetimes of such foils by heating and beam rastering led to marked improvement in some cases,[3] more often only modest increases were obtained.[4] These studies indicated that foil failures were due to radiation induced contraction and subsequent rupture of the foils. Thus, major improvements in foil behavior would more likely be found by modifying the basic structure of the foil to reduce the shrinkage rate.

Several different techniques have been employed to produce carbon foils which have quite different properties from conventional foils. Those being pursued most vigorously at present include vapor deposition on heated substrates,[5] thermal treatment of conventional foils,[6] and glow-discharge cracking of hydrocarbons.[7] The latter technique, used originally to produce hard adherent coatings,[8,9] was extended to thin self supporting foils and has now been used successfully at a number of accelerator laboratories. Further lifetime enhancement has been obtained by providing sufficient slack in the foil to allow some contraction to occur before excessive stresses can develop.[10] By combining these techniques, foil lifetimes have been increased by well over an order of magnitude in many cases.

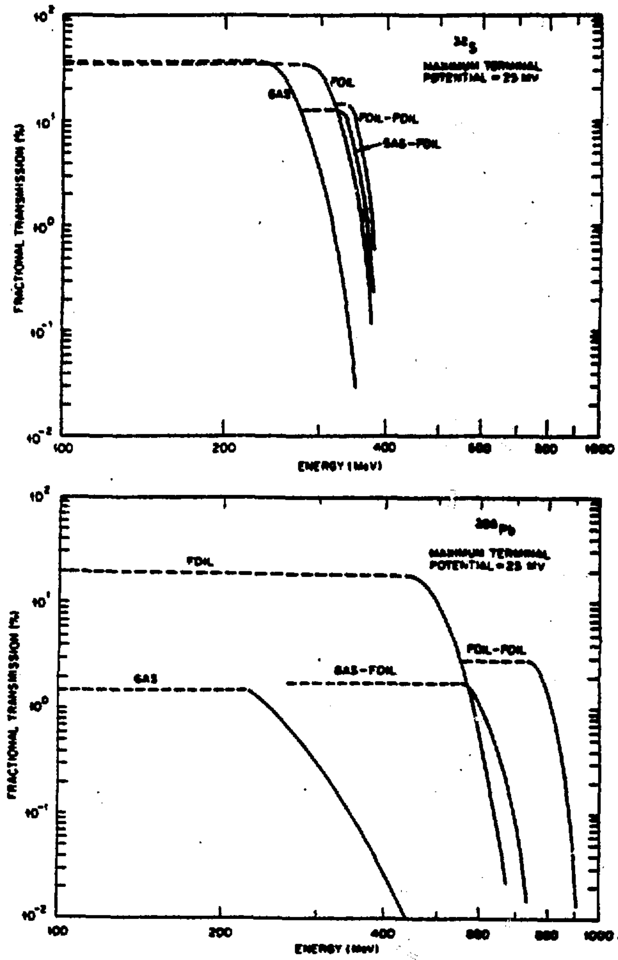

Fig. 1. Fractional transmission (analyzed beam/injected beam) for ^{32}S or ^{208}Pb beams through the HHIRF tandem accelerator for various possible stripper combinations.

Lifetime Measurements

The "lifetime" of a stripper foil is defined herein as the fluence (integrated particle current/cross-sectional area of the beam) required to produce some type of "failure", where the criteria used to define "failure" differ depending on the experimental conditions. For measurements in which the foil condition can be monitored visually, mechanical failure (holes or tears) is usually employed. However, where observation is impractical, such as in the accelerator terminal, failure is typically defined as the point at which the intensity of an analyzed beam has decreased to one-half of the initial value. Therefore, "failure" in the latter case

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may actually result from thickening, inhomogeneities and possibly other effects which could reduce the transmission and quality of the beam. Since the size of the beam is usually unknown in the latter type of measurements, most absolute lifetimes are based on the first criterion. However, since comparative lifetimes for different types of foils are also of interest, relative lifetimes based on both criteria are also important.

The qualitative dependence of foil lifetimes on various foil- and beam-related properties is summarized in table 1. Here it can be seen that to a good approximation, the lifetimes of conventional vapor-deposited foils depend only on the properties of the incident beam, while for foils made by the glow-discharge process, strong dependence on thickness and production details have been reported. Because of the weaker lifetime dependence on foil-specific parameters for vapor deposited foils, they can provide a convenient baseline for evaluating the performance of other types of foils. Several lifetime measurements on vapor deposited foils are given in table 2. Also shown are semi-empirical estimates based on radiation damage theory.[17] This expression reproduces the beam parameter dependence for all but the higher energy ^{127}I data. The latter discrepancies may indicate the onset of a strong temperature dependence since the beam spot was significantly hotter (calculated temperatures of 1100-1300 K) than for the other measurements.

Table 1. Dependence of carbon-foil lifetimes on various foil- and beam-related parameters.

	Parameter	Lifetime-Dependence$^{a)}$: Conventional foils	Lifetime-Dependence$^{a)}$: "Super" foils*
Foil Specific	*Foil Thickness*	Weak$^{b)}$	Strong
	Foil Diameter	Weak	Weak
	Foil Temperature	Medium?$^{c)}$	?
	Production Details$^{d)}$	Weak	Strong
Beam Specific	Beam Energy	Strong	Strong
	A,Z of Beam	Strong$^{e)}$	Strong
	Current Density	Strong	Strong

a) Variations of less than a factor of two (weak), factors of 2 to 3 (medium), or greater than a factor of three (strong) reported for *different values of the indicated parameter.*

b) From ref. 22.

c) Strong dependence reported in ref. 3 was not observed in later measurements.

d) Includes release agents, source material, deposition rates, etc.

e) Weak dependence reported in ref. 23 was not observed in later measurements.

*Based primarily on studies of foils made by the glow-discharge *process. Indications are that the same dependence* holds for other types of long-lived foils.

As noted in table 1, the lifetimes of "super" foils, which collectively includes all those having lifetimes significantly greater than conventional foils, are more complex. For example, those made by glow discharge cracking of hydrocarbons are sensitive to the discharge voltage. The lifetimes are found to drop rapidly for applied potentials less than about 2000V, but become reasonably constant above about 2500V.[25,14] Also, while there is little dependence on the hydrocarbon gas used,[17,25] there does appear to be a dependence on the gas pressure, and hence the power input.[11] Of more significance is the dependence on the foil thickness. The fluences of 10 MeV ^{35}Cl ions required to break various types of foils

Table 2. Lifetimes of conventional vapor-deposited carbon foils.

Incident Beam	E (MeV)	$T(p\mu A \cdot min/mm^2)^{a)}$ Meas.	$T(p\mu A \cdot min/mm^2)^{a)}$ Calc.$^{b)}$	Ref.
^{14}N	0.02	0.021	0.05	17
^{32}S	12-13	≫4.5	2.7	19
^{32}S,^{35}Cl	12	2.6$^{c)}$	2.1,2.7	20
^{35}Cl	10	1.5	1.8	11
^{40}Ar	1.2	0.32	0.17	14,15
^{40}Ar	4.8	1.0	0.67	13
^{127}I	4.9	0.03$^{c)}$	0.025	16
^{127}I	10.5	0.06	0.053	16
^{127}I	50.8	0.8,4.5$^{d)}$	0.26	18
^{127}I	105	>3	0.53	18,24

a) Lifetime to mechanical failure except as noted.

b) From $T(p\mu A \cdot min/mm^2) = 0.0018 \frac{E(ev)}{MZ^2}$ (ref. 17).

c) Lifetime for analyzed beam intensity to decrease by 50%.

d) Strong thickness dependence reported ($T = 0.8$ for 10 $\mu g/cm^2$ foils, $T = 4.5$ for 20 $\mu g/cm^2$ foils) may be due to foil temperature effects.

with thicknesses in the range of 2 to 10 $\mu g/cm^2$ are shown in fig. 2, where the markedly different behavior of glow discharge and vapor deposited foils is quite evident.

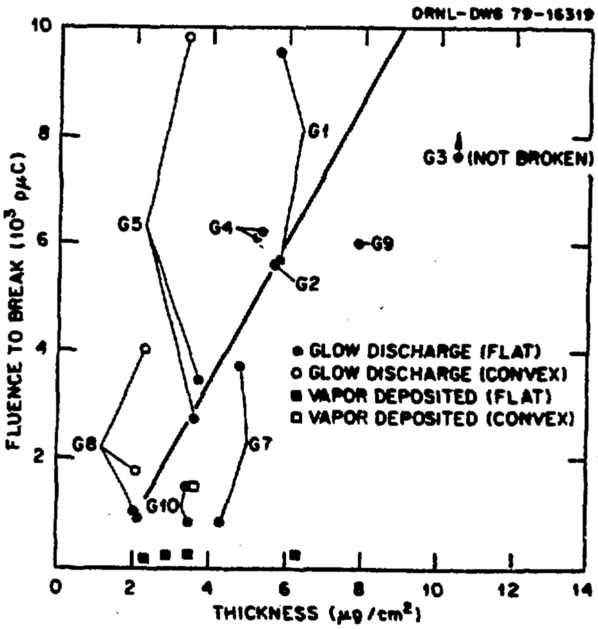

Fig. 2. Thickness dependence of carbon stripper foil lifetimes.

Because "super" foil lifetimes depend on so many parameters, it is only practical to compare absolute lifetimes where all of the pertinent information is known. The data available for long-lived foils are summarized in table 3. The calculated values use the empirical thickness dependence deduced in ref. 17 and is seen to reproduce the measured lifetimes, to better than a factor of two in most cases, for a wide range of beam parameters and foil thicknesses. However, as in the case of conventional foils, there are large differences between the measured and calculated lifetimes for higher energy ^{127}I beams. Again, it is not clear whether this results from a change in the beam energy dependence or from thermal effects.

Table 3. Lifetimes of "super" carbon stripper foils produced by A) glow-discharge method, B) vapor-deposition on heated/treated substrate.

	Incident Beam	E (MeV)	I/A (pμA/mm²)	Thickness (μg/cm²)	T(pμA·min/mm²)[a)] Meas.	T(pμA·min/mm²)[a)] Calc.[b)]	Ref.
A)	^{14}N	0.020	0.03	6	0.64	0.98	17
	^{32}S	12	0.1	5	10.3	39	25
	^{35}Cl	6	0.1	5	9.5[c)]	16	25
	^{35}Cl	10	0.1	2.1	6.6	5.3	11
	^{35}Cl	10	0.1	3.5	14	15	11
	^{35}Cl	10	0.1	5.4	42	29	11
	^{35}Cl	10	0.1	7.9	40	47	11
	^{35}Cl	12	0.1-0.24	5	11[c)]	31	20
	^{40}Ar	1.2	0.07	5-10	3	2.4-5.8	15
	^{40}Ar	1.2	0.07	15-30	10	9.2-19	15
	^{40}Ar	4.8	0.04	15	>28	37	13
	^{127}I	10.5	0.08	≈10	1.3	1.8	16
	^{127}I	50.8	0.1	10,20	0.8,4.5	9,19	18
B)	^{40}Ar	3.5	0.08	5-6	10	8.1	21
	^{40}Ar	3.5	0.08	10	35	17	21
	^{127}I	10.5	0.02	10	0.5	1.8	16

a) Lifetime to mechanical failure, except as noted.

b) From $T(p\mu A \cdot min/mm^2) = [0.0073t(\mu g/cm^2) - 0.010]\,\frac{E(eV)}{MZ^2}$ (ref. 17).

c) Lifetime to half-intensity in analyzed beam.

Additional measurements can be compared by relating the lifetimes to those for conventional foils measured under the same conditions. Enhancement factors for glow discharge and JAERI-type foils are summarized in table 4. It can be seen that the lifetime advantage over conventional foils covers a wide range depending on the thickness of the "super" foils.

Foil lifetimes can also be increased by compensating for the foil contraction by providing slack in the foil at the production stage. This is usually accomplished by mounting the foil on an aluminum ring whose diameter is then reduced by means of a tapered die,[10] although other "slackening" techniques have also been reported.[11,12] Typically, the slackening process allows the foils to contract by approximately 15% in area before stresses are developed. Measured enhancement factors are given in table 5, and appear to be relatively insensitive to the beam parameters. These measurements also covered a wide range of foil thicknesses and no obvious dependence on this parameter could be observed.

Table 4. "Super" foil lifetime enhancement factors (relative to conventional foils).

Incident Beam	E (MeV)	Thickness (μg/cm²)	Enhancement Factors[a)] GD	Enhancement Factors[a)] JF	Enhancement Factors[a)] LT	Ref.
^{35}Cl	10	2.1-3.5	4.2			11
	"	4.3-5.8	20			"
	"	>7.9	>21			"
^{40}Ar	4.8	15	>26			13
^{40}Ar	1.2	5-10	9.1			15
	"	15-30	22			15
^{127}I	10.5	10	24	8.8		16
^{127}I	4.9	10	7.2	4.4		16
^{40}Ar	3.5	5-6		~6		21
	"	10		~80		5
^{32}S	12	5	3.5			25
^{35}Cl	6	5	9.2			25
^{32}S	10.6	5			10	6

a) Lifetime relative to that for conventional (vapor deposited) foils: GD = glow discharge, JF = JAERI-type foil (treated/heated substrate), LT = laser treated.

Table 5. Lifetime enhancement by foil "slackening".[a)]

Incident Beam	E (MeV)	I/A (pμA/mm²)	Enhancement[b)] VDS	Enhancement[b)] GDS	Enhancement[b)] JFS	Ref.
^{35}Cl	10	0.1	5.9	2.9		11
^{40}Ar	1.2	0.07		1.8		15
^{127}I	10.5	0.05		3.7	6.4	16
^{127}I	4.9	0.02		3.9	3.4	16
^{35}Cl	12	0.2		3.0		25
^{35}Cl	6	0.1	7.2	2.0		25
^{40}Ar	4.8	0.04	5			13
		Average:	6.0	2.9	4.9	

a) Foils were slackened by reducing the mount diameter by 7-8%, corresponding to ≈ 15% available "free" area.

b) Lifetime relative to unslackened foil of the same type. VDS = vapor deposited (slackened); GDS = glow discharge (slackened); JFS = JAERI-type foil (slackened).

It has also been reported[25] that the lifetimes of glow-discharge foils are dependent on the beam intensity. Under irradiation with 12 MeV ^{35}Cl beams, the fluence required for mechanical failure of slackened glow-discharge foils decreased from 82 mC at 1.5 μA incident beam, to 47 mC for a 3 μA beam. Such an effect would have serious consequences and the need for further study of beam-intensity dependence is clearly indicated.

In contrast to the sizable enhancements given in table 4, measurements with higher-energy (~ 0.5 MeV/A) ^{127}I beams[18] indicate that unslackened glow discharge foils show no improvement in lifetime over conventional foils. Whether this is a real beam-energy effect or if it is due to subtle differences in production techniques is not known at the present time.

Failure Mechanisms

When used as strippers, factors other than mechanical failure may limit the usable lifetimes of carbon foils. One of the factors to be considered is the change in foil thickness during irradiation. Figure 3 shows the average thickness vs. fluence of several different types of foils under irradiation with 10-MeV ^{35}Cl ions. There are several interesting features to be noted here. For example, the slackened vapor deposited foil has a fairly long lifetime, but increases in thickness by almost a factor of two up to the break point. By contrast, the slackened glow discharge foil, after a rapid initial increase, displays a relatively constant average thickness over most of its lifetime. The measurements on unslackened "super" foils show that the thickness at the later stages of the irradiation can be significantly less than the initial value.

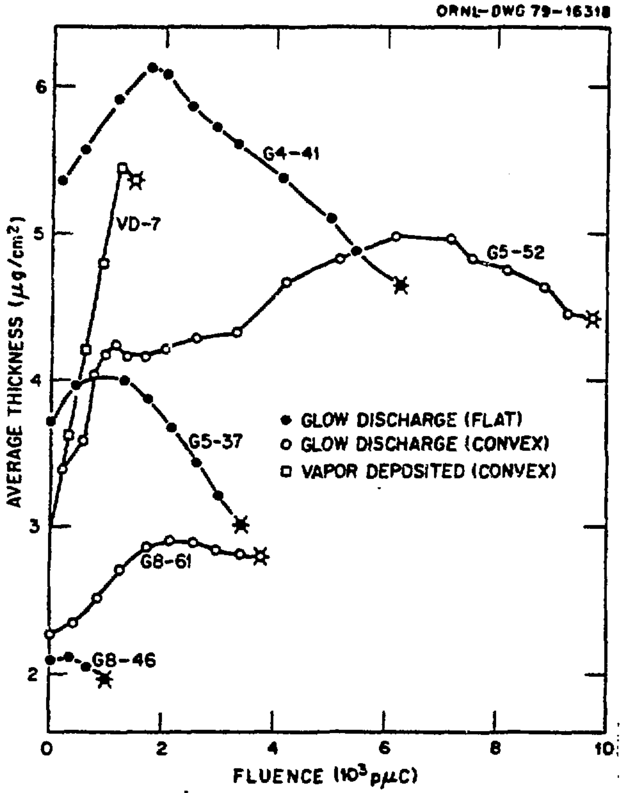

Fig. 3. Thickness changes of several different types of stripper foils, as a function of fluence, irradiated by 10 MeV ^{35}Cl ions.

Low resolution scans[17] of several irradiated foils are shown in fig. 4 and indicate reductions in thickness by 20-25% after irradiation to near the predicted lifetime. However, correcting these values for the finite size of the scanning beam, which has been done by assuming a Gaussian beam intensity distribution, the true reduction in thickness at the center of the irradiated area is approximately twice this value. Thus, the foil thickness at the center of the beam spot has been reduced by almost 50%. Similar results are also reported in higher-resolution scans of a thick (35 μg/cm^2) glow-discharge foil[26] irradiated to ~ 1/3 of the predicted lifetime. This suggests that the thinning rate may be greater for thick foils. The thinning rates observed in these studies were found to be in reasonable agreement with those expected from calculated sputtering rates.[27] These results imply that the incident beam will see a continually increasing macro-inhomogeneity in the stripper foil.

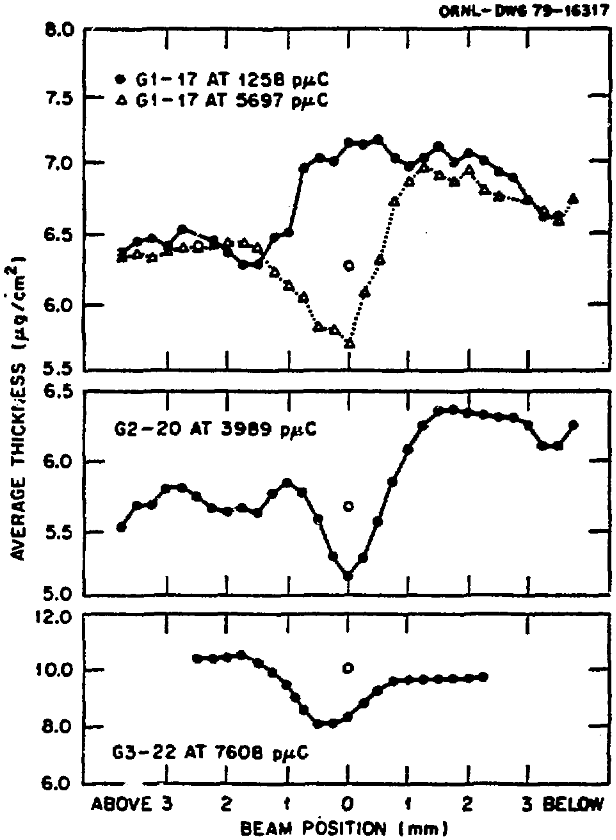

Fig. 4. Thickness scan of irradiated glow-discharge foils.

In addition to macro-inhomogeneities, there is evidence that micro-inhomogeneities, in the form of grains with dimensions on the order of 5 μm, are formed during irradiation of glow discharge foils with 72 MeV ^{79}Br beams.[20] These studies also indicated a continual increase in the average foil thickness, in contrast to the thinning discussed above. This behavior may be related to the small beam spot (0.13 mm^2) employed and the high foil temperature (estimated to be ~ 1000 K) induced by the tightly focused, high current density beam.

Either of these types of inhomogeneities will result in increased energy dispersion in the analyzed beam and might account for the slow decrease in analyzed beam intensity which has been observed with slackened glow-discharge foils.[16,25] Since large changes in intensity and energy resolution will often be intolerable, the useful lifetimes of stripper foils may well be limited by restrictions on analyzed beam properties rather than by mechanical failure of the foil.

Summary

The increased demand for heavy-ion beams from electrostatic accelerators provided the impetus to improve the performance of carbon stripper foils. Significant improvements in radiation-lifetimes have resulted from efforts by several groups to modify the structure of thin carbon foils by a variety of techniques. The production of foils by the glow-discharge cracking of hydrocarbon gases, developed by the Daresbury-Harwell group and now in use at several laboratories around the world, resulted in foil lifetimes ranging from 4 to more than 20 times those of conventional vapor-deposited foils. Similar lifetime improvements are found for foils produced by the heated/treated substrate technique developed at JAERI and for laser-treatment of vapor-deposited foils being pursued at Munich. Further lifetime enhancements, by factors ranging from ~ 3 for glow-discharge foils to ~ 6 for conventional foils, are obtained by the simple but ingenious idea of foil "slackening".

These results have significantly reduced the limitations on electrostatic accelerator operation imposed by stripper foil lifetimes. Even so, the lifetimes become marginal as one goes to higher beam intensities and heavier projectiles. For example, consider a 1 μA injected beam of ^{127}I at 25 MV terminal voltage irradiating a 10 mm^2 area of the stripper foil. Under these conditions, a 5 $\mu g/cm^2$ slackened glow-discharge foil could be expected to survive for approximately 1 hr. Thus, further improvement in foil lifetime would be desirable.

Unfortunately, the outlook for further large increases in carbon-foil lifetimes is not very bright. Thickness measurements on irradiated foils have already shown reductions in foil thickness by as much as 50%. This thinning process, due primarily to sputtering, leads to macro-inhomogeneities in the foil. In addition, there is evidence that micro-inhomogeneities may be formed under certain beam conditions. Either of these processes will result in increased energy dispersion in the analyzed beams and probable rejection of the stripper foil. There are also indications that the lifetimes of conventional and "super" foils are comparable at higher beam energies and/or foil temperatures. Further studies will be required before the behavior of stripper foils in these regimes can be clarified.

Acknowledgments

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